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Calculated Gamma Output from a 6-kilogram Sphere of Neptunium

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Summary

We previously modeled a 6 kg neptunium sphere with pyDMTK 2.0.0b, a python-based intrinsic radiation (INRAD) modeling tool, on the MOONLIGHT machine.¹ Here we report results from version 2.0.1b on SNOW, another TriLab Linux Capacity Cluster (TLCC) resource on the Laboratory's Turquoise network. We also present gamma output from MCNP® 6.2.0² in terms of discrete line strengths, full gamma spectra, and dose rate maps for visualization. Results from both models agree with recent gamma measurements.

Introduction

The Monte Carlo code MCNP® is ideally suited for intrinsic radiation (INRAD) application because of its flexible source and tally features. MCNP's general source definition (SDEF) allows modeling of complex source terms, especially when coupled with MISC,^{3,4} although such models may be computationally expensive and/or require expert users. Tools like pyDMTK¹ provide convenient alternatives to MCNP when rapid results are required. Recent INRAD sources for MCNP, MISC, and pyDMTK application include depleted uranium (DU), highly enriched uranium (HEU), and plutonium. We continue to expand our radioactive source evaluations and promote verification and validation (V&V) of any INRAD modeling software.^{5,6,7,8,9}

Neptunium Sphere

The neptunium sphere (Figures 1 and 2) is a well-documented test article subject to numerous gamma and neutron measurements.^{10,11,12} Gamma measurements on this object were first reported in 2002 and results from a measurement campaign in 2014 are shown in Table 1.^{13,14} We used these data to test our pyDMTK model. Details regarding neptunium decay chain are described by Court.¹⁵ The cited reference also includes gamma spectra from the Np sphere based on MCNP's legacy INRAD option.¹⁶

¹ Shores, Hutchinson, and Miller, Proposed Test Problems for PyDMTK, LA-CP-18-20134 (14 Mar 2018)

² Werner, Ed., MCNP User's Manual—Code Version 6.2, LA-UR-17-29981 (2017); MCNP® and Monte Carlo N-Particle® are registered trademarks owned by Triad National Security, LLC, manager and operator of Los Alamos National Laboratory

³ Solomon, MCNP Intrinsic Source Constructor (MISC): A User's Guide, LA-UR-12-20252 (2012)

⁴ Solomon, The Intrinsic Source Constructor Package: Installation and Use, LA-UR-17-22234 (2017)

⁵ Andrews and Sood, Demonstrating MCNP Correlated Fission Capabilities and MCNP Associated Packages: Intrinsic Source Constructor, MCNPTools, and DRIFT (Detector Response Function Toolkit), LA-UR-16-25779 (2016)

⁶ Shores et al., Predicted radiation output from plutonium and uranium oxide, LA-UR-16-23990 (08 Jun 2016)

⁷ Tucker et al., Simulation of Photon energy Spectra Using MISC, SOURCES, MCNP and GADRAS, LA-UR-12-24107 (2012)

⁸ Rawool-Sullivan, Simulating Authentic Gamma-Ray Spectra: A combined MCNP-GADRAS simulation of an HPGe gamma-ray spectrum collecting using objects containing uranium and plutonium isotopes, LA-CP-11-00765 (2011)

⁹ Rawool-Sullivan, Mattingly, and Mitchell, Use of MCNP + GADRAS in Generating More Realistic Gamma-Ray Spectra for Plutonium and HEU Objects, LA-UR-12-23282 (2012)

¹⁰ Mosteller and Loaiza, Creation of a Simplified Benchmark Model for the Neptunium Sphere Experiment, LA-UR-03-3574, *Trans. Am. Nuc. Soc.*, 89, 624 (2003)

¹¹ Sanchez et al., Criticality of a ²³⁷Np Sphere, LA-UR-03-5267 (2003)

¹² Estes, (U) Preliminary Calculational Studies of a 6 Kg Np Sphere, X-5:GPE-03-106(U), internal LANL memo (10 Nov 2003)

¹³ Moss and Frankle, Gamma-ray Measurements of a 6-kg Neptunium Sphere, LA-UR-02-3601 (2002)

¹⁴ Shores, Hutchinson, et al., LA-CP-14-00470 (24 Apr 2014)

¹⁵ Court, (U) ²³⁷Np Decay-Gamma Spectrum Calculations, X-5:01-08(S), internal LANL memo (16 Apr 2001)

¹⁶ Shelton, Hendricks, and Estes, INRAD: Intrinsic Radiation MCNP, LA-13358 (01 Sep 1997)

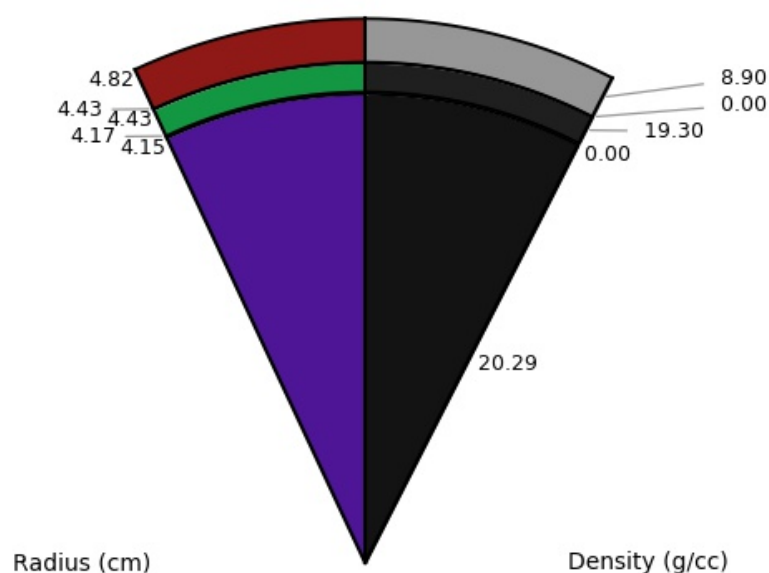


Figure 1. Model of the neptunium sphere as shown within pyDMTK 2.0.1b

FINISHED SPHERE SPECIFICATIONS	
Neptunium mass	6070.4g
Np sphere diameter	8.29cm (3.265in.) neglecting residual sprue blemish
Percent of theoretical density	99.4
Tungsten shield inside diameter	8.331cm (3.280in.)
Tungsten shield out side diameter	8.854cm (3.486in.)
Tungsten shield mass	1082.3g
Inner nickel cladding inside diameter	8.865cm (3.490in.)
Inner nickel cladding outside diameter	9.246cm (3.640in.)
Inner nickel cladding mass	431.6g
Outer nickel cladding inside diameter	9.256cm (3.644in.)
Outer nickel cladding outside diameter	9.637cm (3.794in.)
Outer nickel cladding mass	442.6g
Mass of complete assembly	8026.9g
Bare sphere initial dose rate	2.5R/hr contact
Bare sphere dose rate after grinding	2.0R/hr contact
Sphere dose rate with single nickel cladding layer	650 mR/hr contact
Sphere dose rate with two nickel cladding layers	300 mR/hr contact

Figure 2. Neptunium sphere specifications¹⁷

TABLE 1. Selected gamma results from a measurement campaign and pyDMTK model

E (keV)	Measured Configuration (4pi gps)				pyDMTK Calculation (4pi gps)	Calculation/Exp. ratio (C/E)			
	A	B	C	average		A	B	C	Avg.
300.3	1.06E+7	1.52E+7	1.53E+7	1.37E+7	1.43E+07	1.35	0.94	0.94	1.04
312.2	7.99E+7	1.14E+8	1.14E+8	1.03E+8	1.05E+08	1.31	0.92	0.92	1.02
340.8	1.35E+7	1.92E+7	1.92E+7	1.73E+7	1.94E+07	1.44	1.01	1.01	1.12
375.5	3.31E+6	4.65E+6	4.62E+6	4.19E+6	--	--	--	--	--
398.6	8.67E+6	1.22E+7	1.22E+7	1.10E+7	--	--	--	--	--
415.8	1.28E+7	1.81E+7	1.81E+7	1.63E+7	1.83E+07	1.43	1.01	1.01	1.12

Table 1 configurations A, B, and C refer to a “hot spot” location relative to the HPGe detector. Since measurements are sensitive to this location, the average is also reported; see Shores and Hutchinson for more information regarding the measured system geometry.¹⁴ In addition to full spectral comparisons, calculations (C) and experiments (E) are often considered in terms of individual line strengths. Previously reported C/E line strength ratios were 0.95, 1.03 and 1.06 for the 312, 340 and 415 keV lines, respectively, although that model assumed 100% Np-237 metal and an earlier version of DMTK. We were also comparing to an *average* of three measured configurations.¹⁴ Measurement variations are mainly an artifact of detector location relative to “hot spots” that formed as materials migrated during the casting process.¹⁷

Inspection of Table 1 reveals good agreement with pyDMTK for four passive lines—especially regarding configurations B and C—and this suggests our previous modeling agreement was fortuitous. Since our 2014 effort, we now employ a more realistic neptunium material description as described in the next section. Moreover, we note pyDMTK’s neptunium gamma data were recently modified to be consistent with those from PeakEasy, a Los Alamos gamma-ray spectroscopy software tool.^{18,19}

The pyDMTK results from 2.0.1b are identical to those from version 2.0.0b.¹ We treated two small gaps (between Np/W and W/Ni interfaces) as void and modeled the two nickel layers as a monolithic cladding. The complete pyDMTK model is listed in an output file found in Appendix A.

Additional information regarding this object is found in an International Criticality Safety Benchmark Evaluation Project (ICSBEP) report,²⁰ and efforts toward a new benchmark are underway.²¹ Such a benchmark is important considering variation in reported neutronic results; multiplication of the bare sphere has been reported as 1.9,¹³ 2.48,¹² and 2.5.¹⁴ We find 2.48 here.

¹⁷ Yeamans et al., FABRICATING A TUNGSTEN SHIELDED AND ICKEL CLAD NEPTUNIUM SPHERE, LA-UR-01-3786 (11 Jul 2001); as a historical footnote, one of the authors (EFS) worked in the group responsible for the Np sphere’s shielding design and happened to observe the cast sphere as it emerged from its mold

¹⁸ Mercer and Rooney, PeakEasy, LA-UR-09-06522

¹⁹ Karpus, General Nuclide Identification with PeakEasy, LA-UR-20-30094 (09 Dec 2020)

²⁰ Loaiza NEPTUNIUM-237 SPHERE SURROUNDED BY HEMISPHERICAL SHELLS OF HIGHLY ENRICHED URANIUM, SPEC-MET-FAST-008, Rev 2 (30 Sep 2009)

²¹ Culter et al., Neptunium Subcritical Observation (NeSO) Integral Benchmark Experiment Design, LA-UR 17-29495 (2017)

MCNP Model and Postprocessing

Based on the excellent agreement between pyDMTK models, good agreement with data (Table 1), and fortuitous agreement with previous efforts, it was natural to consider more sophisticated modeling and inter-code comparisons. Thus, we modeled the sphere with MCNP6.2, assuming 20.289 g/cc neptunium and the following atom fractions:²⁰

```
m1  93237.80c 5.0926E-02
    92233.80c 1.8577E-06
    92234.80c 2.9633E-07
    92235.80c 1.4074E-05
    92236.80c 7.8349E-08
    92238.80c 1.5626E-06
    94238.80c 8.2340E-07
    94239.80c 1.6271E-05
    94240.80c 1.1619E-06
    94241.80c 3.1166E-08
    94242.80c 1.6032E-07
    95241.80c 3.3375E-07
    95243.80c 9.1575E-05.
```

As usual for such INRAD applications, each MCNP “model” implies a pair of independent simulations: one for gammas and one for neutrons. The former provides the direct gamma component while the later provides the neutron component *and* those contributions from (n,gamma) reactions.

Each simulation tracked 100 million neutron histories and 5 billion gamma histories.

MCNP output files were parsed with the *mctaltool* utility (designed to extract information from Monte Carlo Tally or MCTAL files) and converted into a “GAM” file with the *gam_convert.pl* utility as shown below. Further details are described by Rawool-Sullivan et al.⁹

```
[eshores@sn-fey2] cat neptunium.sh

/usr/projects/transportapps/local/ml/bin/mctaltool -n 5 -x e -o
gtal_5_100_ENDF6.out endf6.im

/usr/projects/transportapps/local/ml/bin/mctaltool -n 5 -x e -o
ngtal_5_100_ENDF6.out endf6ng.im

/usr/projects/transportapps/local/ml/bin/mctaltool -n 105 -x e -o
ntal_5_100_ENDF6.out endf6ng.im

/usr/bin/perl ./gam_convert.pl -gammafile=gtal_15_100_ENDF6.out -
ngammafile=ngtal_5_100_ENDF6.out -neutronfile=ntal_105_100_ENDF6.out

mv file.gam ENDF6_100cm_ring.gam
```

Descriptive filenames are useful for archiving (and sharing information between organizations), and the above example is based on an ENDF6 data evaluation. This example’s MCNP tallies were made on an axisymmetric ring of 100cm radius centered on the Np sphere, and Solomon’s *mctaltool* was used for parsing MCNP’s output. MCNP users may create their own parsing tools.

Any simulated gamma spectra produced from the above steps must account for the detector response prior to comparison with measured data. This work exercised Sandia National Laboratory's (SNL) Gamma Detector Response and Analysis Software (GADRAS) to produce a synthetic spectrum based on "LANL Detector X."²² GADRAS reads MCNP's output gamma spectra (when placed in GAM file format) as input.

The gamma ray tracing (GRAY) module in pyDMTK directly produced those line strengths reported in Table 1. Line strengths from MCNP6, on the other hand, were inferred from the synthetic spectrum in a manner analogous to measurement interpretation. Gamma leakages for six selected lines are shown in Table 2, and the simulated spectrum is directly compared to a measurement (05260411a.chn) in Figures 3 and 4.²³ Additional details are found in Appendix B.

TABLE 2. Selected Gamma results from a measurement campaign and MCNP6 model

E (kev)	Measured Configuration (4pi gps)				MCNP6 Calculation (4pi gps)	Calculation/Exp. ratio (C/E)			
	A	B	C	average		A	B	C	average
300.3	1.06E+7	1.52E+7	1.53E+7	1.37E+7	1.42E+07	1.34	0.93	0.93	1.04
312.2	7.99E+7	1.14E+8	1.14E+8	1.03E+8	9.09E+07	1.14	0.80	0.80	0.88
340.8	1.35E+7	1.92E+7	1.92E+7	1.73E+7	1.80E+07	1.33	0.94	0.94	1.04
375.5	3.31E+6	4.65E+6	4.62E+6	4.19E+6	4.62E+06	1.40	0.99	1.00	1.10
398.6	8.67E+6	1.22E+7	1.22E+7	1.10E+7	1.03E+07	1.19	0.84	0.84	0.94
415.8	1.28E+7	1.81E+7	1.81E+7	1.63E+7	1.56E+07	1.22	0.86	0.86	0.96

MCNP gamma ray output may be visualized as exposure (Figure 5) or dose rate (Figure 6) maps. Exposure rates were calculated in air using appropriate tally multipliers while dose rates were calculated with fluence-to-dose conversion factors historically used within the Los Alamos INRAD program.²⁴ As expected, the two maps are qualitatively similar.

The health physics community often refers to "contact" dose rates and such values, as the name implies, are meant to articulate dose rates on or near an object's surface. Such measurements are subjective and obviously depend on detector placement. "The final assembled 6kg sphere contact gamma dose rate measured about 300 mR/hr"¹⁷ and our contour plots (Figure 3) indicate 300 mR/hr is approximately 2-3 cm from the cladding surface. Considering the resolution of our dose map, modeling assumptions, and unknown details regarding the contact measurement, this is very good agreement.

²² Horne et al., GADRAS-DRF 18.6 User's Manual, SAND2016-4345 (May 2016)

²³ Hutchinson, Subject: RE: Two Sample GAM files, personal electronic communication to Shores et al. containing multiple measurements and log book entries from 5/26/2004 (12 Jul 2018)

²⁴ Neutron and Gamma-Ray Flux-to-Dose-Rate Factors, American Nuclear Society, ANSI Standard ANSI/ANS-6.1.1-1977 (1977)

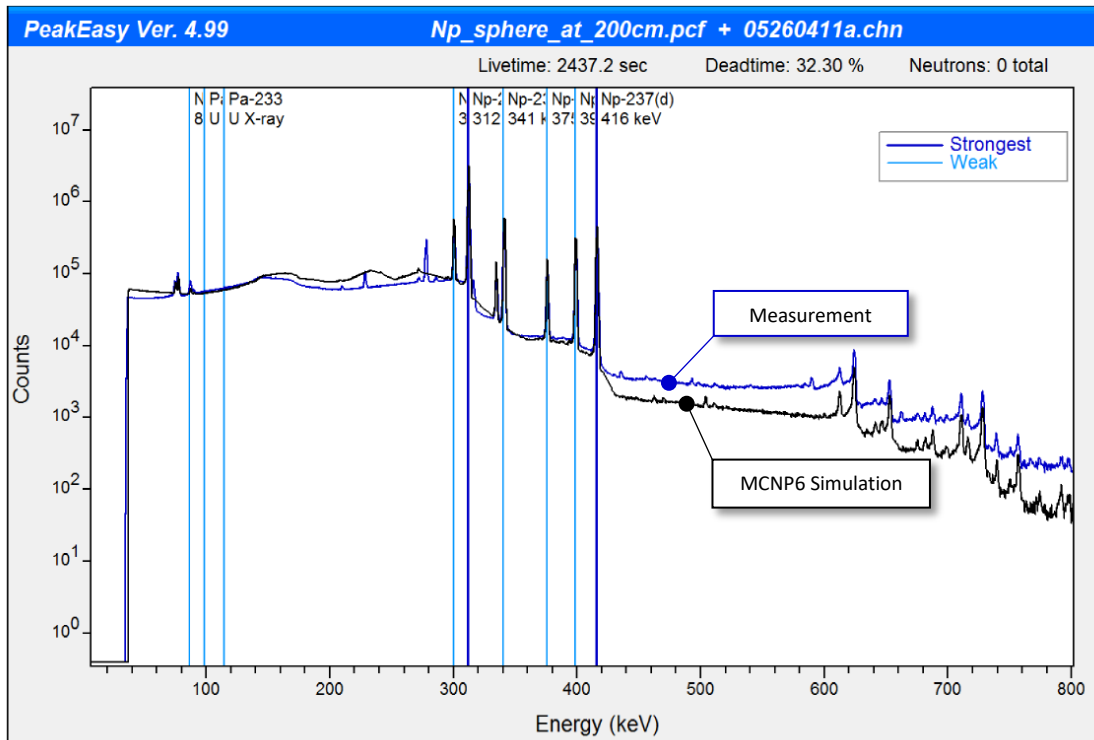


Figure 3. Gamma spectra from the bare neptunium sphere

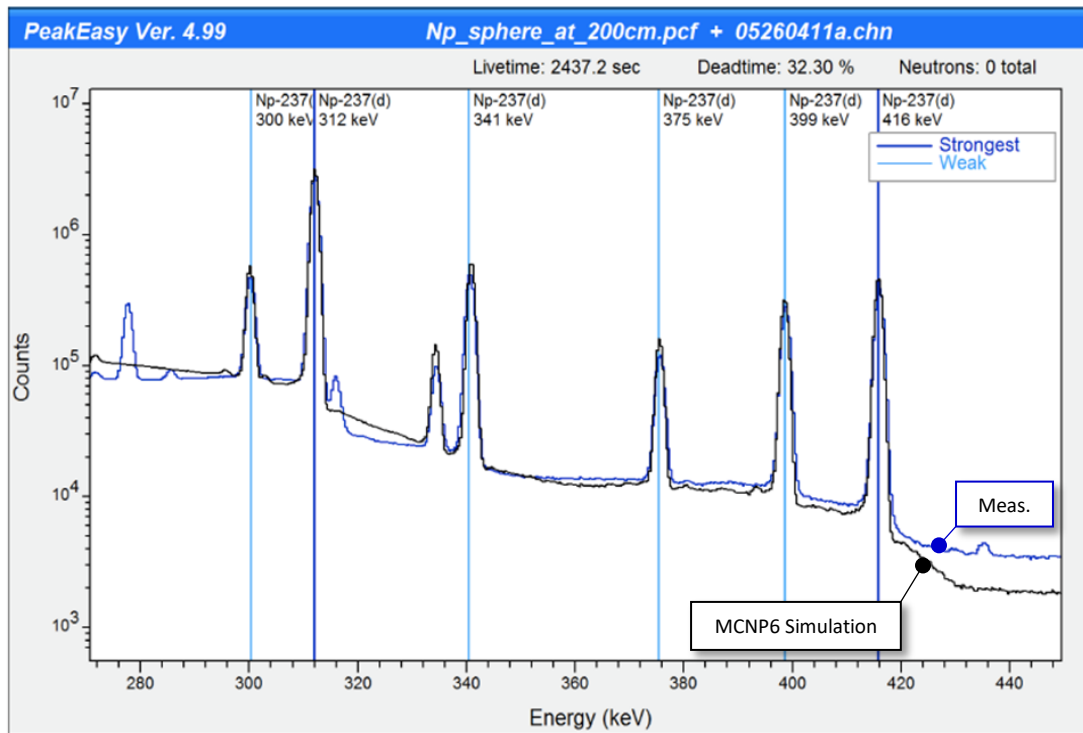


Figure 4. Gamma spectra shown on an expanded energy scale

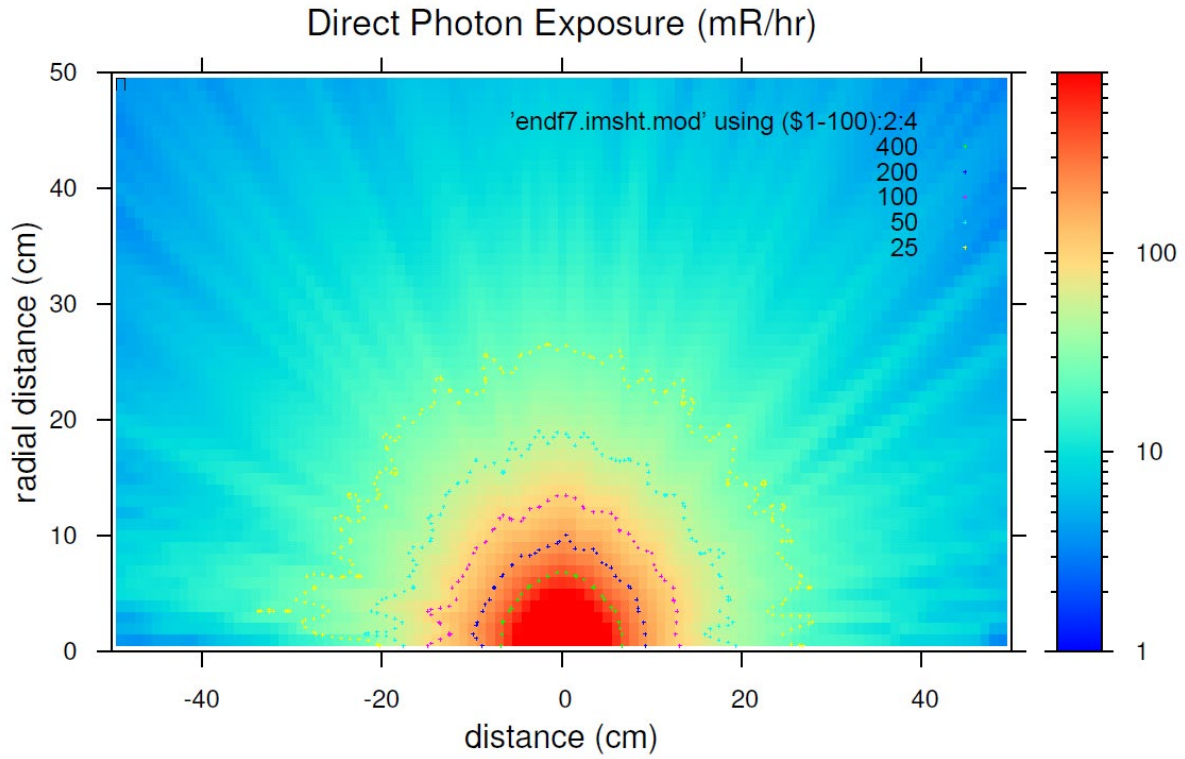


Figure 5. Exposure rate map

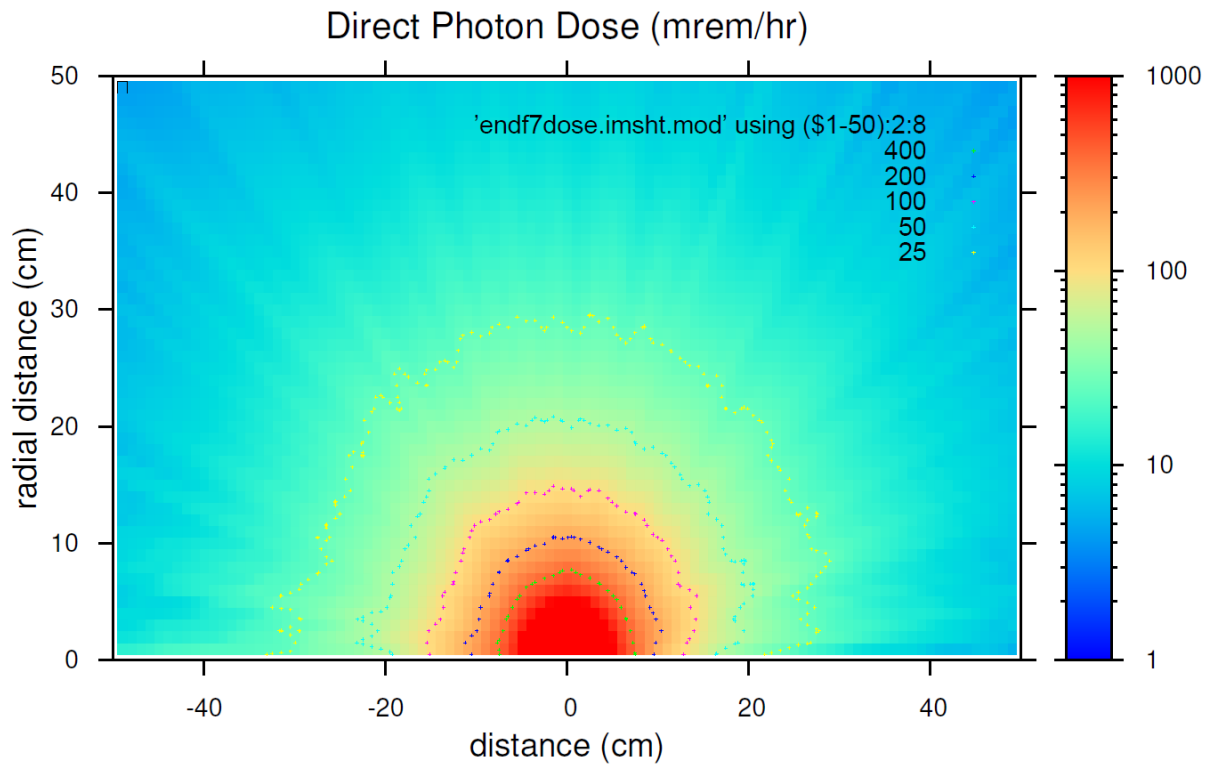


Figure 6. Dose rate map

Neutron leakage

This work's main focus was gamma leakage. Neutron leakage is also reported by pyDMTK, and we briefly discuss that output here. Our neptunium sphere model's multiplication is 2.48. As expected, PARTISN and MCNP report nearly identical k_{eff} results as highlighted in the pyDMTK output listing (Appendix A). Our intrinsic source (444 n/s) is within the range calculated by Estes (441-489 n/s) and, given the system multiplication, leads to pyDMTK's neutron leakage of 788 n/s. This leakage, like that of Estes, is "about a factor of 15 too low" relative to early measurements from the bare sphere (e.g., 1.2×10^4 n/s).¹² A preliminary estimate from the original neptunium isotopic analysis indicated up to 0.0041 g of Cm-244 might be present in the neptunium ball. Indeed, the spontaneous fission rate of Cm-244 ($\sim 1 \times 10^7$ n/g-s) is such that our model leakages are readily increased (e.g., 1×10^4 to 1×10^5 n/s) by adding microgram levels of Cm-244 to our assumed material composition. Addition of light target impurities to the bulk material is another plausible source of neutrons due to (α, n) reactions, but the modest curium addition provides a very simple modeling adjustment.

Conclusions

We modeled a six-kilogram neptunium sphere with pyDMTK and MCNP6 to create intrinsic radiation (INRAD) output for direct comparison with measurements. Full gamma spectra were created with MCNP and detector responses were accounted for with GADRAS. Line strengths from pyDMTK are consistent with those derived from an MCNP generated synthetic spectrum, and results from both models are in good agreement with those derived from a 2014 measurement campaign.

The process of creating synthetic gamma spectra provided:

- another example of MCNP's full spectrum modeling capability when coupled with MISC;
- "GAM" files for direct comparison with GADRAS output and measured spectra;
- dose rate estimates from a significant quantity of special nuclear material;
- an evaluation of pyDMTK software on a new computer platform; and
- another test problem for the pyDMTK user community.

The neptunium sphere model described here is a valuable INRAD test problem and contributes to the ongoing validation of pyDMTK.

Acknowledgements

The authors thank Bev Duran-Cash (GS-NCCP) for supporting this work and continuing to encourage documentation of related efforts. Dean Mitchell (SNL) and Noel Nachtigal (SNL) helped one of authors (EFS) with GADRAS. Steve Myers (NEN-2, Ret.) is gratefully acknowledged for many technical discussions on the matter.

Appendix A. Output from pyDMTK

We executed the following commands on the SNOW platform:

```
[eshores@sn-fey1] module use /usr/projects/packages/DVM/modulefiles
```

```
[eshores@sn-fey1] module load pydmk/2.0.1b
```

```
[eshores@sn-fey1] pyDMTK
```

```
[eshores@sn-fey1] module use /usr/projects/mcnp/modules
```

```
[eshores@sn-fey1] module load mcnp6/6.2
```

```
[eshores@sn-fey1] mpirun -np 128 mcnp6.mpi name=endf7.i
```

Neptunium sphere (model003) output from pyDMTK:

***** DMTK summary for model 'model003' *****

GEOMETRY SUMMARY:

shell	radius (cm)	material	density (g/cc)	volume (cc)	mass (kg)
1	4.149	neptunium_237	20.289	299.17	6.070
2	4.168	void	0.000	4.13	0.000
3	4.427	tungsten	19.300	60.13	1.160
4	4.432	void	0.000	1.23	0.000
5	4.818	nickel	8.902	103.82	0.924

NEUTRON SOURCE SUMMARY:

Absolute source rates (n/s):

Point source at center:
spectrum:
strength: N/A n/s

shell	spont.	fiss. src	(a,n) source	(a,n) interface src
1		4.44e+02	0.00e+00	N/A
2		0.00e+00	0.00e+00	0.00e+00
3		0.00e+00	0.00e+00	0.00e+00
4		0.00e+00	0.00e+00	0.00e+00
5		0.00e+00	0.00e+00	0.00e+00

Total source rate: 4.436e+02 (n/s)

Fractions of total source:

Point source at center: 0.00e+00

shell	spont.	fiss. src	(a,n) source	(a,n) interface src
1		1.00e+00	0.00e+00	N/A
2		0.00e+00	0.00e+00	0.00e+00
3		0.00e+00	0.00e+00	0.00e+00
4		0.00e+00	0.00e+00	0.00e+00
5		0.00e+00	0.00e+00	0.00e+00

MEASURED DATA COMPARISONS:

Passive Gamma Comparisons (4pi-leakage gam./s):

Isotope	Line (keV)	Measured	RE %	GRAY Calc.	C/M	# sigma dev.
U-233	146	N/A	N/A	5.21e-04	N/A	N/A
U-233	291	N/A	N/A	4.72e+00	N/A	N/A
U-233	317	N/A	N/A	1.14e+01	N/A	N/A
U-235	144	N/A	N/A	1.03e-03	N/A	N/A
U-235	186	N/A	N/A	9.51e-01	N/A	N/A
U-235	205	N/A	N/A	3.16e-01	N/A	N/A
U-238	258	N/A	N/A	8.29e-04	N/A	N/A
U-238	743	N/A	N/A	6.71e-02	N/A	N/A
U-238	766	N/A	N/A	2.43e-01	N/A	N/A
U-238	1001	N/A	N/A	1.03e+00	N/A	N/A

Measured	RE %	Partisn	C/M	# sigma dev.	MCNP	RE %	C/M	# sigma dev.
N/A	N/A	7.88e+02	N/A	N/A	N/A	N/A	N/A	N/A

~~Partisn = N/A~~
~~MCNP = N/A~~

Measured		Partisn			MCNP		
M	RE %	M = 1/(1-k)	C/M	# sigma dev.	M = 1/(1-k)	C/M	# sigma dev.
2.50	1.00	2.48	9.94e-01	6.24e-01	N/A	N/A	N/A

Partisn = 0.5975
MCNP = 0.5970

[illegible]

Appendix B. Energy spectrum examples

Energy spectra from two MCNP6 simulations are shown below. Here we compare problem *endf7.im* (08 Jul 2018) to *endf7toy4.im* in Figures B1. The *endf7toy* series (March 2021) has modified tally bins relative to the default bin structure expected by GADRAS.

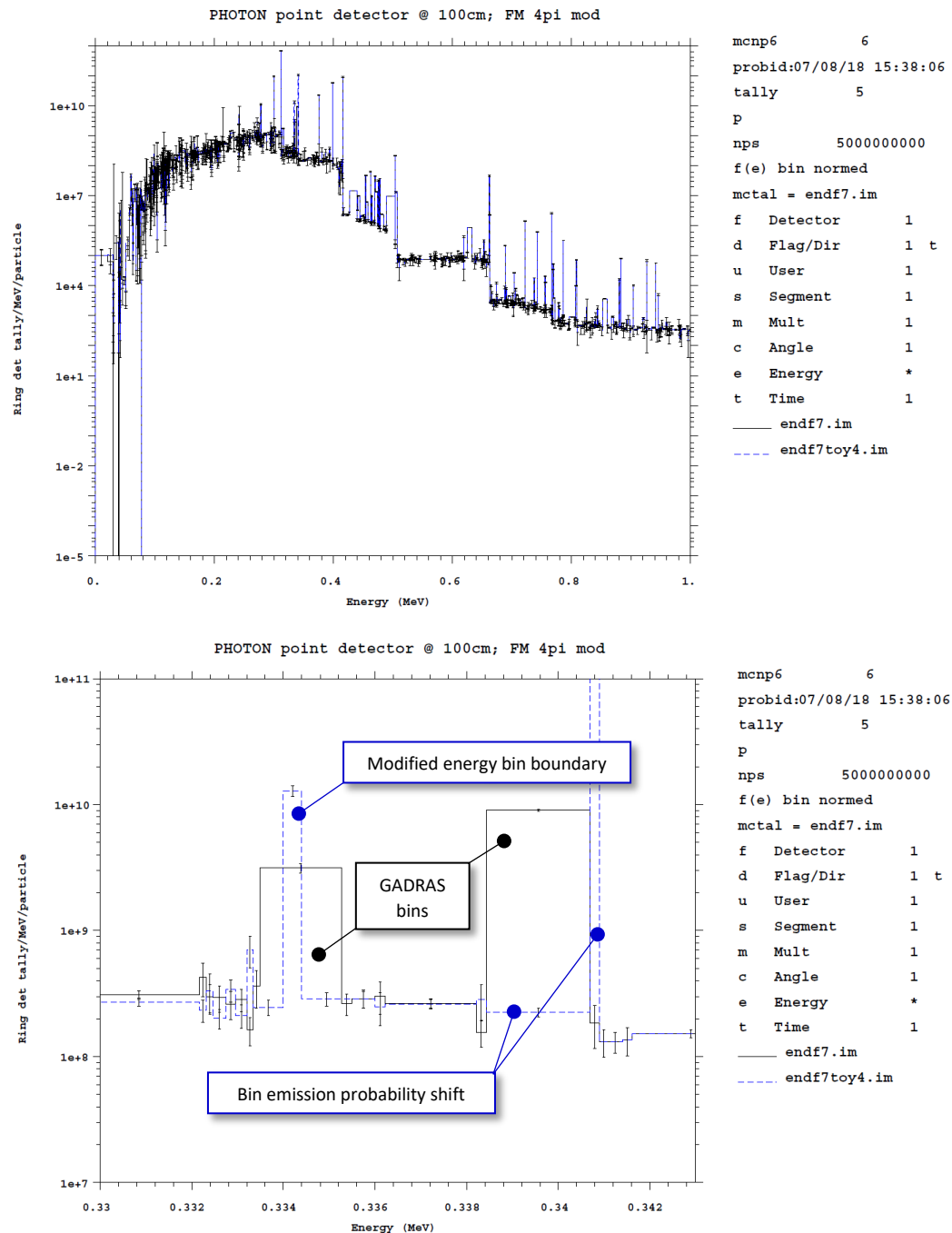


Figure B1. Gamma leakage from the Np sphere; 0-1 MeV (top) and 0.33-0.343 MeV (bottom)

The bottom panel of Figure B1 reveals the difference between the two MCNP simulations: shrinking the boundaries of that bin containing the 334 keV peak at the expense of adjacent bins. Folding the MCNP6 leakage spectra with a GADRAS detector response results in Figure B2. Here, the pair of relatively coarse bins shown above effectively wash out that pair of peaks that should exist near 341 keV and produce a broad hump (e.g., region near 340 keV in Figure B2).

Rather than adjust boundaries for that bin containing the 341 keV peak, we retained the GADRAS-specified structure and instead adjusted source emission probabilities within the MCNP simulation. As expected, shifting the peak's emission probability into the immediately adjacent (higher) bin resulted in a subtle peak misalignment as shown in Figure B3.

In short, there are two mechanisms to resolve the pair of peaks hidden within Figure B2's broad hump: shifting bin emission probabilities and modifying individual bin widths. Given the format of the GAM file and defined energy bins, we did not explore the simple addition of more bins in the region of interest. A snippet of the default energy structure from an MCNP input file is shown as Figure B4 (also refer to Figure B1's bottom panel). The source description reveals energy (mev), bin emission probabilities, and uniform biasing for sampling efficiency. If the GAM file expected by GADRAS could accommodate additional energy bins from the MCNP forward model, they would be located in the region labeled **<peak here>**.

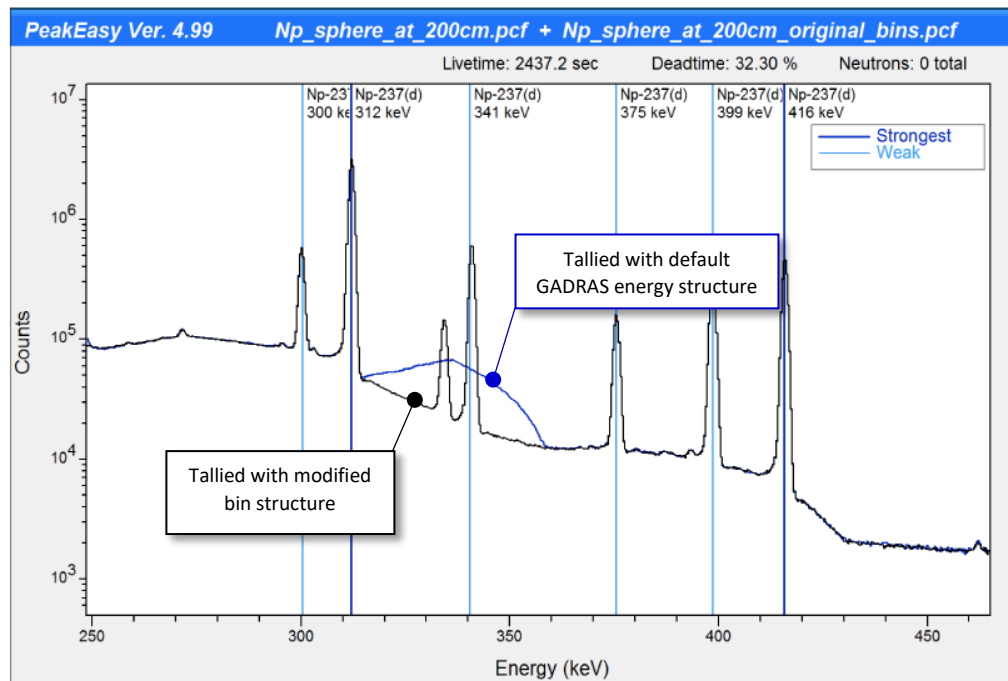


Figure B2. Synthetic MCNP-GADRAS energy spectra tallied on two different bin structures

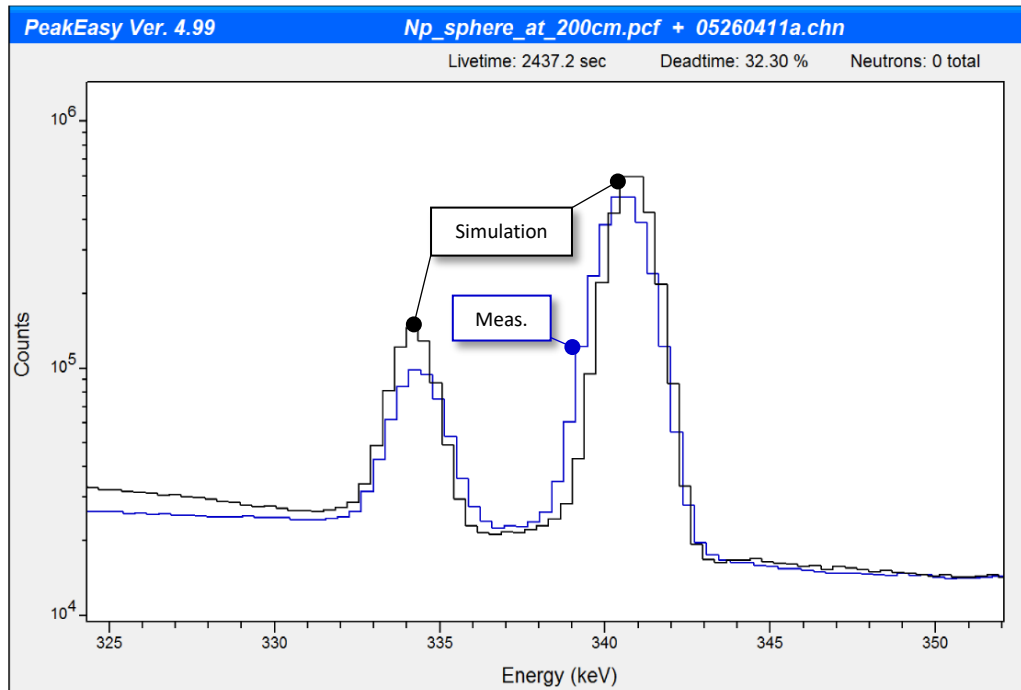


Figure B3. Synthetic and measured gamma spectra near the 334 and 341 keV peaks

Snippet of the MCNP source definition (*sdef*) from MISC (SI, SP, and SB entries):

3.3391e-01	2.2909e-14	1.0000e+00
3.3431e-01	2.7294e-03	1.0000e+00
3.3440e-01	2.2918e-13	1.0000e+00
...
3.3980e-01	3.2078e-16	1.0000e+00
3.4045e-01	3.3810e-12	1.0000e+00
3.4050e-01	1.1385e-02	1.0000e+00
3.4056e-01	3.6151e-10	1.0000e+00
3.4077e-01	1.6004e-13	1.0000e+00
3.4097e-01	8.3423e-20	1.0000e+00

Energy bins from the MCNP tally (mev):

0.31189	0.31206	0.31228	0.31421	0.31443	0.3163	0.31646
0.31662	0.31999	0.32021	0.32075	0.32097	0.32154	0.32176
0.32373	0.32395	0.32411	0.32433	0.32734	0.32756	0.32789
0.32811	0.32931	0.32953	0.33216	0.33232	0.33247	0.33274
0.33296	0.33320	0.33334	0.33348	<peak here>	0.33527	0.33549
0.33622	0.33821	0.33843	<peak here>	0.3407	0.3409	0.34109
				0.3414		

Figure B4. MCNP source and tally regions near the 334 and 341 keV peaks